

**REMARKS**

Applicants acknowledge receipt of an Office Action dated December 5, 2002. In this response, Applicants have cancelled all pending claims (i.e. claims 2-8, 10-12 and 15-25) without prejudice or disclaimer, and Applicants have added claims 26-49. Support for these amendments may be found in the specification as discussed in more detail *infra*. Following entry of these amendments, claims 26-49 are pending in the application.

Reconsideration of the present application is respectfully requested in view of the foregoing amendments and the remarks which follow.

**Interview on March 12, 2003**

Applicants acknowledge, with appreciation, the courtesies extended by Examiner Turner to Mr. Kawamura, Mr. Yasuda, Mr. Schwaab and Mr. Strain during a personal interview on March 12, 2003. During the interview, the outstanding rejections and cited references were discussed in light of newly proposed claims which focused on the relationship between polar groups at the surface of hard carbon-based films and oiliness agents present in a lubricating oil contacting the surface. As discussed during the interview, Applicants are submitting herewith a copy of the article entitled "Diamond and diamond-like phases" from Diamond and Related Materials, Vol. 1, pages 61-62 (1991). As can be seen in Figures 1 and 2, amorphous carbon, graphite and diamond have atomic fractions of hydrogen which are practically 0.

**Rejections Under 35 U.S.C. §103**

On page 2 of the Office Action, the PTO has rejected claims 2-8, 10-12, 15-16 and 18-21 under 35 U.S.C. §103(a) as being unpatentable over U.S. Patent 5,205,188 to Repenning *et al.* (hereafter "Repenning") or JP 6-294307 to Sumitomo (hereafter "Sumitomo") in view of U.S. Patent 5,843,571 to Sho (hereafter "Sho"). In addition, on page 3 of the Office Action, the PTO has rejected claims 2-6, 8, 10-12, 15-16 and 18-21 under 35 U.S.C. §103(a) as being unpatentable over Repenning or Sumitomo in view of U.S. Patent 5,466,431 to Dorfmann *et al.* (hereafter "Dorfmann"). In this response, Applicants have cancelled claims 2-8, 10-12 and 15-25 without prejudice or

disclaimer. Applicants expressly reserve the right to pursue the subject matter of these claims in the present application or a subsequent application, based on the disclosure in this application. In view of the cancellation of claims 2-8, 10-12 and 15-25, Applicants submit that the outstanding rejections under 35 U.S.C. §103 are now moot.

#### **Newly Added Claims**

In this response, Applicants have cancelled all previously pending claims without prejudice or disclaimer and have added claims 26-49.

The presently claimed invention relates to an unexpected beneficial and synergistic relationship between the presence of large numbers of polar groups on the surface of certain hard carbon-based films and oiliness agents present in lubricating oils for internal combustion engines. The present inventors found that the interaction between the oiliness agents and the polar groups on the surface of the hard carbon based films resulted in a surprising reduction in the friction coefficient of the film. The references cited in the outstanding Office Action, taken either individually or in fair combination, fail to teach or suggest this unexpected benefit resulting from the interaction of oiliness agents and polar groups in the surface of a hard carbon-based film. The new claims have been added to focus prosecution on this beneficial and unexpected relationship.

As an initial matter, Applicants note that one of the inventors, Mr. Yasuda, gave a presentation related to the unexpected benefit resulting from the interaction of oiliness agents and polar groups in the surface of hard carbon-based films at an SAE conference in March 2003. A copy of the slides from the presentation and accompanying comments have been submitted herewith.

The data set forth in the presentation (see, for example, slides 6-10, 13, 16 and 18-21) demonstrate the unexpected reduction in friction coefficient realized by the combination of a low hydrogen content (e.g. a large number of polar groups) and oiliness agents, and, with particular regard to slide 8, provide a helpful depiction of the interaction of oiliness agents and surfaces with and without polar groups. Slides 6 and 7 show comparative data from "pin-on-disk" friction tests with (slide 6) and without (slide 7) lubrication. As can be seen in slide 7, the physical vapor deposition - diamond

like carbon ("PVD-DLC") film exhibited friction coefficient at or below 0.07 while the plasma-enhanced chemical vapor deposition ("PE-CVD") diamond-like carbon ("DLC") film exhibited a coefficient of about 0.11. Slide 9 shows that diamond-like carbon films produced by arc-ion plating have almost zero hydrogen and exhibit low friction coefficients under lubricated conditions. Slide 18 shows the relationship between hydrogen content on a surface and contact angle of a water drop (an indication of the relative polarity/surface energy of a surface). The PVD-DLC film has a significantly lower hydrogen content and exhibits a lower contact angle than the PE-CVD film. Thus, it appears that the number of polar groups/surface energy of the PVD-DLC film is higher. Finally, slide 21 demonstrates that the combination of an a PVD-DLC film with a lubricant and oiliness agent produce unexpectedly lower friction coefficients than a PE-CVD DLC film in combination with the same lubricant and oiliness agent.

With regard to claim 26, none of the references cited in the outstanding Office Action, taken either individually or in combination, teach or properly suggest the combination of a "lubricating oil...containing oiliness agents," "a substrate selected from the group consisting of silicon nitride and metal" and "a hard carbon-based film...having been applied by a process that produces in the surface of the film a sufficient number of polar groups to adsorb the oiliness agents from the lubricating oil to a degree to produce a lowered coefficient of friction" recited in claim 26.

In the outstanding Office Action, the PTO maintained obviousness rejections based on (i) the combination of Repenning or Sumitomo and Dorfmann (the "Dorfmann rejection") and (ii) the combination of Repenning or Sumitomo and Sho (the "Sho rejection"). Neither of these rejections properly applies to claim 26 for the reasons set forth below.

To establish *prima facie* obviousness of a claimed invention, all the claim limitations must be taught or suggested by the prior art. *In re Royka*, 180 USPQ 580 (CCPA 1974). See MPEP §2143.03. Here, none of the references relied on in the Dorfmann rejection, taken either individually or in combination, teach or properly suggest a substrate selected from the group consisting of silicon nitride and metal together with the presently claimed hard carbon-based film as recited in claim 26.

Applicants note that the PTO did not apply the Dorfmann rejection to claim 7 in the outstanding Office Action.

Further, none of the references relied upon in either the Dorfmann or the Sho rejection, taken either individually or in combination, teach or properly suggest the specific combination of polar groups on a surface and oiliness agents in a lubricant oil.

In the outstanding Office Action, the PTO has cited Repenning as a reference which relates to a slidable member in a valve operating system. The PTO has alternatively relied on Sumitomo for the disclosure of an adjusting shim which has a surface layer formed of diamond making contact with a cam. The PTO then combined each of these references with a secondary reference, Sho.

Sho relates to a slidable member of a mechanical part which includes an amorphous hard carbon film deposited on a substrate. The PTO relies on Sho for Sho's reference to the use of the slidable member in a wear test in combination HFC-134a + PAG oil (or refrigerating machine oil) with a high pressure friction machine (See Sho, Test Example 2).

The HFC134a + PAG oil of Sho, as described in paragraph 5 of the 132 Declaration submitted with Applicants previous response, is substantively different from the "lubricating oil for an internal combustion engine" as recited in amended independent claims 10, 15, 16, 17 and 18. In this regard, Applicants note that the HFC-134a + PAG oil does not contain an oiliness agent which would be adsorbed at the surface of the hard carbon film disposed on the substrate. As a result, it would be impossible to obtain the low coefficient of friction of the presently claimed invention by using the HFC- 134a + PAG oil. As set forth in paragraph 5 of the 132 Declaration, HFC-134a + PAG oil is a compressor oil. HFC-134a designates a coolant, and PAG means polyalkylene glycol (water-soluble synthetic base oil). Thus, the HFC-134a + PAG oil is a mixture of the coolant and the water-soluble synthetic base oil and, therefore, is significantly different than a lubricating oil for an internal combustion engine which is hydrophobic and contains an oiliness agent. An example of the ratio of the coolant and the water-soluble synthetic base oil in the HFC-134 + PAG mixture is 600 g :160 g. It will be understood that both the coolant and polyalkylene glycol may

not contain an oiliness agent. Thus, Sho taken either individually or in fair combination with either Repenning or Sumitomo, fails to teach or properly suggest the combination of polar groups at the surface of the presently claimed hard carbon-based film and a lubricant oil comprising oiliness agents.

Applicants therefore submit that the Dorfmann rejection and the Sho rejection each fail to establish *prima facie* obviousness of the embodiment of the presently claimed invention set forth in newly added independent claim 26 or the claims that ultimately depend therefrom.

Even assuming *arguendo* that the PTO had established a *prima facie* case of obviousness based on the Dorfmann rejection or the Sho rejection, Applicants submit that the unexpected interaction between polar groups on the surface of the presently claimed hard carbon-based films and oiliness agents present in a lubricant oil in contact with the surface of the hard carbon-based film would be sufficient to rebut that *prima facie* case of obviousness. This unexpected interaction results in a decrease in friction coefficient as can be seen in the data presented in slides 6, 7, 9, 10, 13, 16 and 18-21 of Mr. Yasuda's presentation (see discussion *supra*).

The remarks set forth *supra* with regard to claim 26 apply equally to independent claims 33, 47 and 48.

With regard to claim 33, none of the references cited in the outstanding Office Action, taken either individually or in combination, teach or properly suggest the combination of a "metal or silicon nitride substrate," "an oiliness agent," and "a hard carbon based film comprising a sufficient number of polar groups to adsorb the oiliness agent to a degree to produce a lowered coefficient of friction" as recited in claim 33.

With regard to claim 47, none of the references cited in the outstanding Office Action, taken either individually or in combination, teach or properly suggest the combination of a "metal or silicon nitride substrate," "a hard carbon-based film formed on the substrate, the hard carbon based film having been applied by a process that produces in a surface of the film an increased concentration of polar groups," and "an agent which adsorbs to the surface of the hard carbon-based film as a result of the

presence of the polar groups and which thereby decreases the friction coefficient of the surface" as recited in claim 47.

With regard to claim 48, none of the references cited in the outstanding Office Action, taken either individually or in combination, teach or properly suggest the combination of a "metal or silicon nitride substrate having a film formed thereon," "a lubricant comprising an oiliness agent in contact with the film," and "a hard carbon-based film formed on the substrate, the hard carbon-based film having (i) a surface roughness of not higher than 0.1  $\mu\text{m}$ , (ii) a thickness of 1 to 10  $\mu\text{m}$ , and (iii) comprising a sufficient number of polar groups to adsorb the oiliness agent to produce a coefficient of friction, Ra, of not higher than 0.07" as recited in claim 48.

#### **Support for Claims 26-49**

Support for newly added claims 26-49 may be found in the disclosure when taken as a whole.

#### Oiliness Agents

With particular regard to the phrase "oiliness agent," Applicants note that a search of the Lexis U.S. Patent Database, which includes the text of U.S. Patents from 1790-the present, turned up more than 1000 U.S. Patents which use the phrase "oiliness agent" or "oiliness agents". Forty-seven of these U.S. Patents use the phrase "oiliness agent" or "oiliness agents" in the claims. A listing of these 47 patents has been submitted herewith. Applicants note that this listing includes patents that issued as early as 1937 and as recently as 2002.

As one example, Applicants submit herewith U.S. Patent 4,776,967, which relates to a multi grade engine oil composition that can be used as an internal combustion engine oil. This patent uses the term "oiliness agents" in the claims and provides the following exemplary description: "As the oiliness agent, higher fatty acids such as oleic acid and stearic acid; higher alcohols such as oleyl alcohol; amines; esters; chlorinated fats and oils; and the like can be used."

In addition, Applicants have submitted herewith a copy of pages 398-403 of Thorpe's Dictionary of Applied Chemistry, Fourth Edition, published in 1946, which includes discussions of "The Theory of Boundary Lubrication," "Theories of Oiliness," and on page 402, under the sub-heading "(e) Anti-Friction Additives," a discussion of "Oiliness Compounds":

These substances, exemplified by the long-chain polar compounds present in fatty or vegetable oils, are able to lower the boundary friction and improve the ease and spreading, when present either alone or as solutions in mineral oils. For example, stearic and other long-chain fatty acids are often added to improve lubricants for slow running mechanisms, whilst their metal soaps are often used in wire-drawing, etc.

In view of the foregoing, Applicants submit that a person skilled in the art would understand the phrase "oiliness agent" as used in the present specification to refer to the class of materials belonging to this very old, art-recognized category of lubricant additives, which include boundary lubricants with a polar component (such as the carboxylic acid group present on stearic acid) and a long chain hydrocarbon (such as the  $\text{CH}_3(\text{CH}_2)_{16}$  tail present on stearic acid).

#### Polar Groups

With particular regard to the presence of polar groups on the surface of the hard carbon-based film, Applicants submit that the present disclosure, when viewed in its entirety, clearly communicates to the person skilled in this art an unexpected beneficial interaction between polar groups present on the surface of hard carbon-based films and oiliness agents. The Applicants have provided two exemplary types of films that contain large amounts of polar groups (as compared to other films) and thereby provide for increased adsorption of oiliness agents contained in lubricating oil for an internal combustion engine.

As described in the first full paragraph on page 6 of the specification, hard carbon-based films which comprise nitrogen and oxygen in an amount ranging from 0.5 to 30 at% have a large, e.g. increased, amount of polar groups present on the surface as compared to other films. As a result, oiliness agents can adsorb at the surface thereby lowering the friction coefficient of the surface.

In a parallel discussion in the first full paragraph on page 8 of the specification, Applicants described an alternative mechanism for increasing the number of polar groups present on the surface of hard carbon-based films, namely, decreasing the surface hydrogen content. The text of this paragraph is reproduced below:

Otherwise, the low friction characteristics in lubricating oil can be obtained by controlling the content or concentration of hydrogen at a level of not more than 10 at% in the surface section of the hard carbon-based film. Although measurement of content of hydrogen in the surface section is difficult, the content can be readily estimated from conditions where formation of the hard carbon-based film is accomplished. Accordingly, such a low content of hydrogen can be realized by forming the hard carbon-based film of amorphous carbon by a carbon ion beam process or the like in which hydrocarbon plasma is not used at least during formation of the hard carbon-based film, or by forming the hard carbon-based film of a diamond polycrystal by a thermal chemical vapor deposition (CVD) process. Additionally, such a low content of hydrogen can be realized by forming the hard carbon-based film by an ion plating process, or by a sputtering process. With the thus formed hard carbon-based film, a large amount of polar groups are present at the surface of the hard carbon-based film, and therefore oiliness agents contained in lubricating oil are liable to be physically or chemically adsorbed at the surface of the hard carbon-based film. It will be understood that hydrogen may be contained in the above-mentioned amount in the hard carbon-based film other than the surface section.

In this passage, Applicants acknowledge the difficulties of measuring surface hydrogen content and suggest that the hydrogen content can be estimated from the conditions under which the hard carbon-based film is formed. The Applicants went on to describe a number of methods which one skilled in the art would recognize as methods that would result in the formation of low (e.g. not more than 10%) but preferably near zero hydrogen at the surface (e.g. the diamond polycrystal produced by thermal CVD)<sup>1</sup>. Applicants then state that the "thus formed", (i.e. formed by the processes listed in lines 7-15 which one skilled in the art would recognize as processes which would result in films having low to almost no hydrogen at the surface) hard carbon-based film

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<sup>1</sup> Without careful control or other extraordinary measures, there are practically no compositions that have surface hydrogen contents between 10% and near 0%.



exhibits a sufficiently large amount of polar groups present on the surface for oiliness agents to adsorb at the surface.

The Examples show the use of O<sub>2</sub> and N<sub>2</sub> alone, reduction of H<sub>2</sub> alone and use of combined systems. This shows that the inventors and persons skilled in the art understood that the concept of enhancing the number of polar groups is a generic concept and that both adding O<sub>2</sub> and N<sub>2</sub> as well as reducing the H<sub>2</sub> content are alternative measures for increasing the number of polar groups.

One skilled in the art, taking the present disclosure in its entirety, and considering the specific teachings at page 6, page 8 and the Examples, would view the first full paragraph on page 8 of the disclosure as (1) a recognition of the relationship between the presence of hydrogen on the surface of the hard carbon-based films and the presence of polar groups and (2) a suggestion to maximize polar groups by minimizing the presence of hydrogen based by selecting a method known to result in low to no hydrogen at the surface of hard carbon-based films.

Finally, Applicants note that the corresponding German application has issued as a patent (DE 100 17 459) on March 28, 2002, and claims 1 and 10 have a scope essentially equivalent to the newly added claims. A copy of the issued German patent has been submitted herewith.

**CONCLUSION**

In view of the foregoing amendments and remarks, Applicants respectfully submit that all of the pending claims are now in condition for allowance. An early notice to this effect is earnestly solicited. If there are any questions regarding the application, the Examiner is invited to contact the undersigned at the number below.

Respectfully submitted,

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